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Estimates of the Concentration of Mercury in Bluegill
(*Lepomis macrochirus*) in the Lower End of Watts Bar Reservoir from 1946
to 1988 and Potential Health Risks to Humans

B. G. Blaylock, M. L. Frank, C. R. Olsen, and R. R. Turner

ABSTRACT

The purpose of this study was to predict the concentration of mercury that existed in the edible tissue of bluegill from the lower end of Watts Bar Reservoir between 1946 and 1988 and to determine whether the consumption of bluegill from this area in previous years presented a health risk. Sediment cores were taken from four sites in the reservoir and depth increments were analyzed for ^{137}Cs . Sedimentation rates were determined by relating the increment with the highest concentration of ^{137}Cs to the year (1956) in which high levels of ^{137}Cs were released from the Department of Energy's Oak Ridge Facilities. The depth of the ^{137}Cs peak indicates the rate of sedimentation. The sediment increments, water, and bluegill tissue were analyzed for mercury. A distribution coefficient (K_d) based on current measurements was used to calculate the concentration of dissolved mercury in water during past years. A concentration factor (CF) (fish/water) and concentration ratio (CR) (fish/sediment) were determined for one collection site. This CF and CR were used to calculate the concentration of mercury in bluegill tissue for previous years.

Highest concentrations of mercury in bluegill tissue were predicted for the years 1956-1960 for a collection site near Tennessee River Mile (TRM) 567.5. The average predicted concentration of mercury in these fish for this five-year period was $1.5 \mu\text{g/g}$. Because a mercury concentration of $1.5 \mu\text{g/g}$ exceeds the current Food and Drug Administration action level of $1.0 \mu\text{g/g}$ for fish, calculations were performed to determine whether consumption of bluegill with a mercury concentration of $1.5 \mu\text{g/g}$ would present a health problem.

The average freshwater fish consumption rate for adults surveyed in the U.S. during 1973-1974 was 3.41 kg/yr (9.3 g/day) (Rupp 1980). An average adult who eats fish containing 1.5 μg of mercury per gram would have a mercury intake of 14.0 $\mu\text{g/day}$. This is below the acceptable daily intake (ADI) of 21 μg recommended by EPA (Lappenbusch 1988). The maximum consumption rate reported by Rupp (1980) for the Tennessee region was 24 kg/yr (67.5 g/day) which would provide a daily intake of 101 μg . This level of mercury is above EPA's recommended ADI but less than the 300 $\mu\text{g/day}$ required to produce signs and symptoms of mercury poisoning (Federal Register 1979).

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The release of nonradiological pollutants from the U.S. Department of Energy's Oak Ridge Facilities received little attention until 1982 when it became public knowledge that large amounts of mercury had been released into East Fork Poplar Creek (EFPC) from the Y-12 facilities. EFPC is a tributary of Poplar Creek which flows into the Clinch River arm of Watts Bar Reservoir (Figure 1). The mercury release resulted from a crash program initiated in 1953, a production-scale process "COLEX" or the separation of isotopes of lithium that was started in 1955 and stopped in 1963, after having produced the strategic material needed for national defense. One phase of this process was a solution of lithium in mercury, a lithium amalgam. Many millions of pounds of mercury were essential to the project; and, as a result, large quantities of mercury were lost to EFPC. Estimates of the amount of mercury released into EFPC range from 10^5 to 2.1×10^5 kg (Oak Ridge Y-12 Plant 1983) with the highest discharges occurring during 1957 and 1958. According to Turner et al. (1985) and Loar et al. (1988), most of the mercury released from the Y-12 Plant can still be found in the sediment of Watts Bar Reservoir.

Operations and waste disposal activities on the Oak Ridge Reservation have also introduced ^{137}Cs , a radioactive isotope with a 30-y half-life, into local streams that ultimately drain into the Tennessee River - Watts Bar Reservoir system. Cesium-137 is readily sorbed onto suspended particles and sediments and, as a result, accumulates in sediments. Highest discharges of ^{137}Cs into the river system occurred during 1956 in association with the draining of White Oak Lake at Oak Ridge National Laboratory (ORNL). Sediment core samples taken from Watts Bar Reservoir by Turner et al. (1984) have shown that the elevated level of ^{137}Cs discharged in 1956 is reflected by a peak of ^{137}Cs in the vertical profile of the

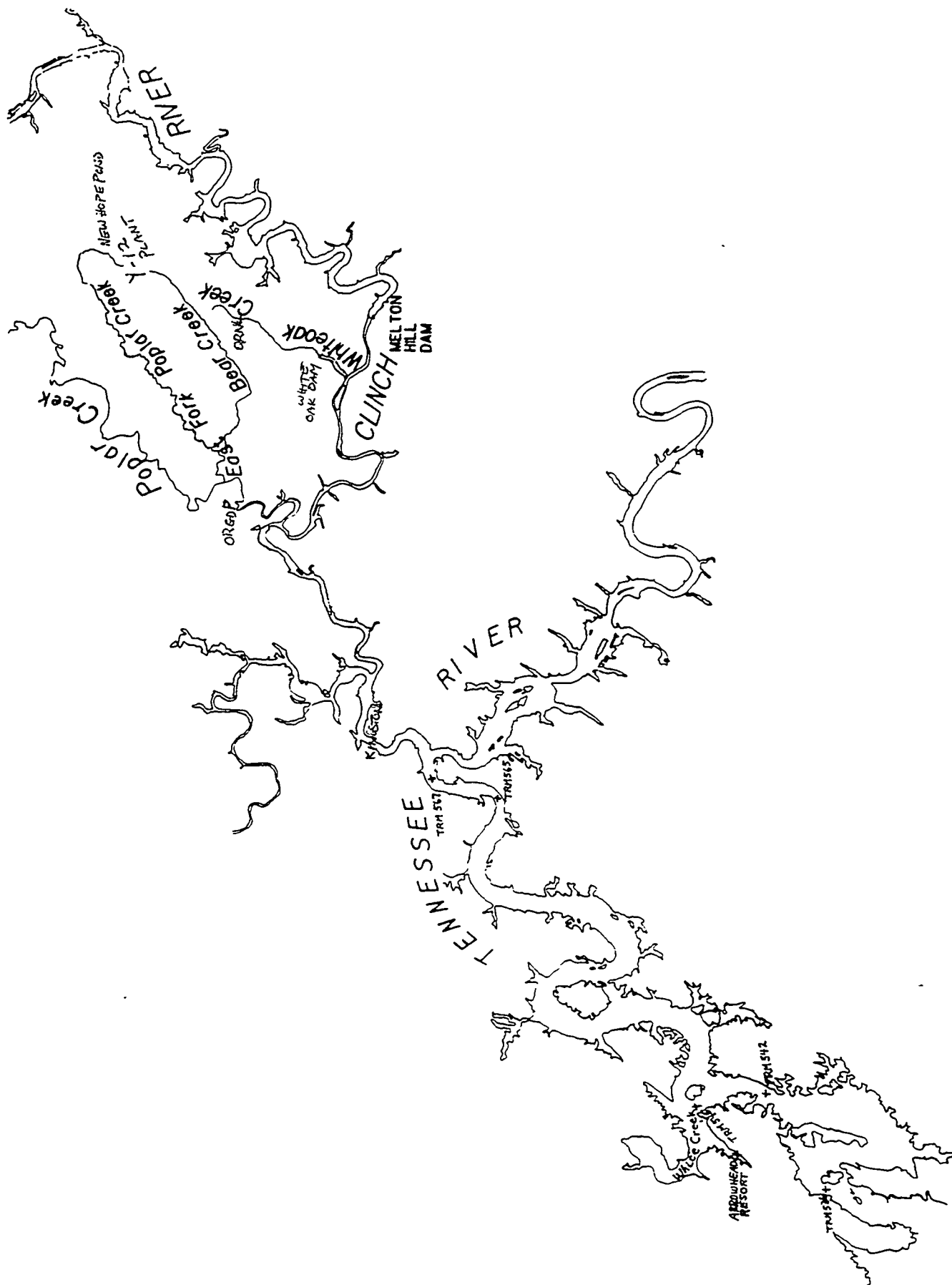


Figure 1. Map showing the Tennessee and Clinch River arms of Watts Bar Reservoir and the lower section of Melton Hill Reservoir.

sediment. By determining the depth below the sediment surface at which the ^{137}Cs peak occurs, the rate of sedimentation can be determined. The sedimentation rate can then be used to date the sediment layers. Data on the vertical distribution of mercury together with the sedimentation rate can be used to estimate the concentration of mercury in surface sediments during previous years. Furthermore, if the concentration of mercury in the sediment and the relationship between the dissolved and particulate phases of mercury in the water column are known, the concentration of dissolved mercury in the water column for previous years can also be predicted.

Because it was not public knowledge that large quantities of mercury had been released into the Tennessee River - Watts Bar Reservoir system until 1982, no data on the concentration of mercury in fish from the reservoir were collected during the time when high levels of mercury were being released. However, by using estimates of past concentrations of mercury in the sediment and/or water, it is possible to estimate the concentration of mercury that would have been in fish during the same period. These estimates can then be used to determine whether mercury in Watts Bar Reservoir fish ever posed a threat to human health.

Purpose:

The purpose of the present study was to:

- (1) predict the concentration of mercury that was in the surface sediment and water of the lower end of Watts Bar Reservoir during the years 1955-1989.
- (2) predict past concentrations of mercury in bluegill from the lower end of Watts Bar Reservoir using both a concentration factor (CF) (fish/water) and concentration ratio (CR) (fish/sediment).

- (3) compare CFs and CRs for mercury in fish from other aquatic environments (North Fork Holston River in Tennessee and the Wabigoon-English River system, Ontario, Canada) with those from Watts Bar Reservoir.

Background:

Concentration factor (fish/water)

The assimilation of a pollutant in aquatic biota is calculated by using a single empirical relationship to represent the transfer of the pollutant from water to organism. This dimensionless transfer coefficient is known as a concentration factor. The CF of an organism or tissue is the ratio of the contaminant in the organism or tissue to that in water:

$$CF = \frac{C_B}{C_w}$$

where C_B is the contaminant concentration in fresh weight of the biota or tissue and C_w is the contaminant concentration in water in equivalent units. The assimilation of a contaminant in aquatic biota is a complex phenomenon, especially in organisms that absorb contaminants directly from water as well as from food via the gastrointestinal tract. The ratio of the equilibrium concentration of the assimilated contaminant in the organism or tissue to the equilibrium concentration of that contaminant in water during the history of exposure is the true CF. A variety of environmental factors can influence the assimilation of contaminants and thus the CF in aquatic organisms (Blaylock 1982).

Concentration ratio (fish/sediment)

Another method for predicting the concentration of pollutants in an organism, although not commonly used, is a concentration ratio (CR) that relates the concentration of a pollutant in an organism to that in the sediment. It is generally accepted that in a stream or lake the concentration of a contaminant in sediment will vary more from location to location than will the concentration in water; therefore, when the number

of samples is limited, the range of predicted concentrations in organisms based on sediment concentrations should vary more than the range of predicted concentrations based on equilibrium water concentrations for the same environment. However, sediment concentrations better reflect the exposure history and show less temporal variation than water. In this study, a comparison of the predicted concentration of mercury in fish inhabiting Watts Bar Reservoir was made using both CF and CR values. Data from studies at other sites are also included to show how site-specific CF and CR values can vary.

Because of the many environmental variables that can influence the CF and CR ratios, it is advisable, when data are available, to use site-specific values for predicting the concentration of a contaminant in fish rather than generic values. For example, the generic CF for ^{137}Cs in freshwater fish is 2000; however, in White Oak Lake located on the Oak Ridge reservation, the site-specific CF is about 400 (Kolehmainen and Nelson 1969).

Method:

Sample collection

Sediment cores from the lower end of Watts Bar Reservoir (Figure 1) were obtained using a Wildco K-B heavy duty gravity corer with a 2-inch stainless steel core barrel and a polycarbonate plastic liner. This device was loaded with a clean barrel and lowered by winch to a height of about 10 feet above the sediment water interface. The winch was then unlocked and the corer allowed to free-fall into the sediment. Duplicate cores were collected at each site, although only one core per site was ultimately retained for analyses.

Sediment cores were returned intact to the laboratory for extruding and sectioning. To minimize mixing of the sediment prior to extrusion, the cores were maintained upright during transit to the laboratory. The cores were extruded and sectioned into either 4-cm intervals for the entire length or 3-cm intervals for the upper 15 cm and 5-cm intervals to the core bottom. Core sections were placed in plastic lined aluminum cans (new 'pudding cups') and sealed with a mechanical canner. Each can was then counted

for gamma ray emitters using a germanium detector and peak height analyzer (Nuclear Data Corp). Geometry correction factors had been previously established for these cans and these factors allowed conversion of raw counts to picocuries of ^{137}Cs . After gamma counting, the lids were partially removed from each can and the contents dried for 16 h in a forced air oven set at 60°C (in accordance with USEPA protocol for handling of sediment samples for mercury analysis). Each can was then weighed to obtain the dry weight of sediment. The dried sediment was pulverized manually using a porcelain mortar and pestle to assure a homogeneous sample for mercury analysis. The mortar and pestle were thoroughly cleaned and dried between each sample to avoid cross contamination.

The pulverized samples were submitted to the ORNL Analytical Chemistry Division (ACD) for analysis of total mercury using ACD Preparation Method 10915 and ACD Analytical Method 1214922. In addition to routine quality assurance/quality control procedures used by the ACD, analysis of a Standard Reference Material (NBS SRM 1646, Estuarine Sediment) was requested to be run with one batch of core samples. The results (0.065 and 0.067 $\mu\text{g/g}$, respectively, for two portions) were found to be in good agreement with the certified value (0.063 ± 0.012 $\mu\text{g/g}$) for this material. Established chain-of-custody procedures were followed from the field collection of the cores through the analysis phase.

To quantify the mercury particle-to-water distribution ratio, a large-volume water sample (approximately 800 L) was collected about 2 m below the water surface near the mouth of White Creek in Watts Bar Reservoir near Tennessee River Mile (TRM) 545. Suspended matter (> 0.45 μm) was removed from the water by continuous-flow centrifugation within 4 h after sample collection. After the suspended matter was dried and weighed, it was analyzed for total mercury by the ACD.

Two 500-mL water samples were collected at the same time and location as the large-volume sample to measure the concentration of dissolved mercury in the water column. The samples were filtered through 0.2- μ m filters and the filtrates were analyzed for total mercury by the ACD.

Bluegill (*Lepomis macrochirus*) were collected from three sites in the lower end of Watts Bar Reservoir (TRM 542, 545, and 565) March 09, 1989. Fillets from eight fish from each location were analyzed for total mercury (Table A1). Chain of custody was maintained for the samples and the Environmental Protection Agency's (EPA) approved modified method 7471 (EPA 1986) was used for the analysis of mercury in the fillets. Quality assurance was maintained throughout the collection and analysis of the samples.

Estimating historical concentrations of mercury

Because sorption onto suspended particles and sediments is the principal mechanism by which many chemically reactive contaminants (such as ^{137}Cs and mercury) are removed from aquatic systems, the history of ^{137}Cs and mercury contamination in Watts Bar Reservoir has been recorded in the sediments. By assuming that the peak concentration of ^{137}Cs in the sediments represents 1956, the sediment layers can be dated and the history of mercury contamination can be documented. As expected, all of the cores show subsurface peaks in mercury and ^{137}Cs (Table A2) that are coincident with their peak discharge histories; however, the sediment depth of the subsurface peaks and the thickness of contaminated sediment vary with location in the reservoir and depend on the rate of sediment accumulation.

Historical estimates for the amount of dissolved mercury in the Watts Bar Reservoir depend on site-specific information concerning the partitioning of mercury between dissolved and particulate phases. This partitioning is expressed quantitatively by assigning it a value or distribution coefficient (K_d), defined as:

$$K_d = \frac{C_p}{C_w}$$

where C_p is the concentration of a specific contaminant associated with a given weight of particles and C_w is the concentration of the contaminant in an equal weight of water. Ideally, this ratio is a measure of the reversible equilibrium partitioning of a contaminant between dissolved and particulate phases and would be a constant. Because most natural environments (including Watts Bar Reservoir) are affected by short-term physical, chemical, and biological processes, chemical equilibrium is continually adjusting.

The particle-to-water distribution for mercury listed in Table 1 was obtained from the 800-L water sample taken in Watts Bar Reservoir near the mouth of White Creek. A particle-to-water ratio of 1×10^5 indicates that the dissolved concentration of mercury is about 100,000 times lower than the concentration on suspended particles and surface sediments. If we assume that the particle-to-water distribution ratio for mercury has been relatively constant at this site throughout the past, then we can estimate past levels of dissolved mercury from the dated levels of mercury in the sediment cores (Table A2).

Table 1. Distribution of mercury between particulate and aqueous phases.

Location & Date of Sample	Hg on Suspended Matter ($\mu\text{g/g}$)	Dissolved Hg ($\mu\text{g/L}$)	Distribution Coefficient
White Creek Mouth (March 09, 1989)	0.51	0.004	1.3×10^5

The concentration of mercury in fish in Watts Bar Reservoir from 1955 to 1988 was predicted by using the estimated concentrations of mercury in water and sediment for those years and multiplying the concentrations by either a CF or CR value, respectively. To obtain a site specific CF and CR for fish from Watts Bar Reservoir, the average concentration of mercury in fish tissues from TRM 545 was divided by the measured concentration of mercury in water and surface sediment (0-4 cm) collected at approximately the same location. Maximum and minimum CF and CR values were obtained by using the maximum and minimum concentrations of mercury measured in the fish tissue.

Results:

Site specific CFs and CRs

The average concentrations for mercury in bluegill for three locations in Watts Bar Reservoir, along with the calculated CFs and CRs, are given in Table 2. The size of the bluegill ranged in length from 13.4 to 18.4 cm (5.3 to 7.2 in) with an average length of about 15 cm (5.9 in). The weight of the bluegill ranged from 47.2 to 96.2 g (1.7 to 3.4 oz) with an average weight of approximately 60 g (2.1 oz). Scales from five bluegill were analyzed to estimate the age of the fish. The ages ranged from 4 to 6 y. Data on individual fish are given in Table A1.

The average measured concentration of mercury in bluegill from TRM 565 was a factor of two greater than that of fish from sites TRM 542 and 545 (Table 2). In comparison, the average measured concentration of mercury in sediment from this area (TRM 567) was greater by a factor of three (Table 2). The higher mercury concentration in sediment at TRM 567 is attributed to the influence of higher mercury concentrations in the Clinch River water before it is diluted with the water of the Tennessee River. Concentrations of mercury in water, fish, and sediment, which are needed to calculate site-specific CFs and CRs, were obtained for only one site (TRM 545); however, mercury concentrations in sediment and fish were obtained for other sites (Tables 2 and A2).

Table 2. Concentration factors and concentration ratios for Watts Bar Reservoir bluegill.

Tenn. River Mile	<u>Tissue Hg</u>		Hg in Water ^b (µg/g)	<u>Fish/Water Conc Factor</u>			Sediment Hg (µg/g)	<u>Fish/Sediment Conc Ratio</u>		
	Ave ^a (µg/g)	Range (µg/g)		ave	max	min		ave	max	min
542	0.030	.017--.054								
545	0.030	.020--.044	0.000004	7500	11000	5000	0.52	0.058	0.085	0.038
565	0.062	.021--.125					1.52	0.041	0.082	0.014

^a Eight bluegill per site.

^b Hg dissolved in water.

The CF calculated for bluegill collected on March 09, 1989, for location TRM 545 averaged 7,500 and ranged from 5,000 to 11,000 (Table 2). The CFs were determined by using the measured concentration of dissolved mercury ($0.004 \mu\text{g/L}$) in water and the measured concentrations of mercury in fish. Maximum and minimum CFs were calculated for the highest and lowest concentrations measured in bluegill from TRM 545. The sediment CR for TRM 545 averaged 0.058 and ranged from 0.038 to 0.085; the average CR for TRM 565 was 0.041 and ranged from 0.014 to 0.082 (Table 2).

Predicted concentrations of mercury in bluegill for different locations in Watts Bar Reservoir during previous years based on predicted water concentrations are listed in Tables 3, 4, 5, and 6. A sediment distribution coefficient (K_d) of 10^5 , determined from measured concentrations of mercury in water and surface sediment at TRM 545, was used to predict the previous concentrations of dissolved mercury in water at the other locations and depths. Concentrations of mercury in bluegill for previous years were estimated by using the site specific CF from TRM 545 together with predicted dissolved mercury concentrations in water. Higher predicted mercury concentrations in bluegill at TRM 567.5 reflect the higher measured concentrations in sediment core samples from this site. Thus, it follows that higher predicted concentrations of mercury in fish are directly related to higher concentrations in sediment. As the concentration of mercury in surface sediment decreased from the peak years (1955-1960) to the present, the predicted concentrations of mercury in fish also decreased.

A comparison of the predicted concentrations of mercury in fish in Watts Bar Reservoir using the CF value of 7500 determined for TRM 545 and the average CR value of 0.058 for TRM 545 and 567 are given in Table A2. A comparison of the results shows that the predicted tissue concentration based on the water concentration is always higher than the predicted tissue concentration based on sediment values. This difference is an artifact resulting from using 10^5 rather than 1.3×10^5 (Table 1) as the K_d and applying the CF and CR obtained with TRM 545 data to the other sites.

Table 3. Predicted concentrations of mercury in tissue of Watts Bar Reservoir bluegill for Tennessee River Mile 545 based on predicted concentrations of dissolved mercury in water.

Historical Time Period (Year)	Predicted Hg Conc in H ₂ O ^a (µg/g)	Predicted Hg Conc in Bluegill Tissue		
		Average (µg/g)	Maximum (µg/g)	Minimum (µg/g)
1984-1986	0.000005	0.037	0.055	0.025
1981-1983	0.000005	0.037	0.055	0.025
1978-1980	0.000005	0.037	0.055	0.025
1975-1977	0.000006	0.045	0.066	0.030
1972-1974	0.000007	0.052	0.077	0.035
1969-1971	0.000008	0.060	0.088	0.040
1966-1968	0.000011	0.082	0.121	0.055
1964-1965	0.000018	0.135	0.198	0.090
1961-1963	0.000023	0.172	0.253	0.115
1958-1960	0.000047	0.352	0.517	0.235
1956-1958	0.000055	0.412	0.605	0.275
1953-1955	0.000011	0.082	0.121	0.055
1950-1952	0.000005	0.037	0.055	0.025
1947-1949	0.000003	0.022	0.033	0.15
1944-1946	0.000001	0.007	0.011	0.005
		0.007	0.011	0.005

^a Dissolved Hg, estimated using a particle-to-water distribution ratio (K_d) of 10^5 .

Table 4. Predicted concentrations of mercury in tissue of Watts Bar Reservoir bluegill at White Creek Mile 1.3 (TRM 545) based on predicted concentrations of dissolved mercury in water.

Historical Time Period (Year)	Predicted Hg Conc in H ₂ O ^a (µg/g)	Predicted Hg Conc in Bluegill Tissue		
		Average (µg/g)	Maximum (µg/g)	Minimum (µg/g)
1988-1989	0.000005	0.037	0.055	0.025
1987	0.000006	0.045	0.066	0.030
1986	0.000005	0.037	0.055	0.025
1985	0.000005	0.037	0.055	0.025
1984	0.000006	0.045	0.066	0.030
1981-1983	0.000006	0.045	0.066	0.030
1979-1980	0.000007	0.052	0.077	0.035
1977-1978	0.000007	0.052	0.077	0.035
1975-1976	0.000009	0.067	0.099	0.045
1973-1974	0.000010	0.075	0.110	0.050
1971-1972	0.000011	0.082	0.121	0.055
1969-1970	0.000012	0.090	0.132	0.060
1967-1968	0.000015	0.112	0.165	0.075
1965-1966	0.000016	0.120	0.176	0.080
1963-1964	0.000024	0.180	0.264	0.120
1961-1962	0.000027	0.202	0.297	0.135
1959-1960	0.000050	0.375	0.550	0.250
1957-1958	0.000080	0.600	0.880	0.400
1955-1956	0.000065	0.487	0.715	0.325
1953-1954	0.000004	0.030	0.044	0.020
1951-1952	0.000002	0.015	0.022	0.010
1949-1950	0.000002	0.015	0.022	0.010

^a Dissolved Hg, estimated using a particle-to-water distribution ratio (K_d) of 10^5 .

Table 5. Predicted concentrations of mercury in tissue of Watts Bar Reservoir bluegill for Tennessee River Mile 567.5 based on predicted concentrations of dissolved mercury in water.

Historical Time Period (Year)	Predicted Hg Conc in H ₂ O ^a (µg/g)	Predicted Hg Conc in Bluegill Tissue		
		Average (µg/g)	Maximum (µg/g)	Minimum (µg/g)
1986	0.000015	0.112	0.165	0.075
1984-1985	0.000014	0.105	0.154	0.070
1983	0.000018	0.135	0.198	0.090
1981-1982	0.000024	0.180	0.264	0.120
1980	0.000032	0.240	0.352	0.160
1978-1979	0.000023	0.172	0.253	0.115
1977	0.000023	0.172	0.253	0.115
1975-1976	0.000033	0.247	0.363	0.165
1974	0.000034	0.255	0.374	0.170
1972-1973	0.000030	0.225	0.330	0.150
1969-1971	0.000029	0.217	0.319	0.145
1966-1968	0.000041	0.307	0.451	0.205
1963-1965	0.000051	0.382	0.561	0.255
1960-1962	0.000068	0.510	0.748	0.340
1957-1959	0.000244	1.830	2.684	1.220
1955-1956	0.000190	1.425	2.090	0.950
1952-1954	0.000063	0.472	0.693	0.315
1950-1951	0.000012	0.090	0.132	0.060
1948-1949	0.000004	0.030	0.044	0.020
1946-1947	0.000004	0.030	0.044	0.020

^a Dissolved Hg, estimated using a particle-to-water distribution ratio (K_d) of 10^5 .

Table 6. Predicted concentrations of mercury in tissue of Watts Bar Reservoir bluegill at Tennessee River Mile 536.5 based on predicted concentrations of dissolved mercury in water.

Historical Time Period (Year)	Predicted Hg Conc in H ₂ O ^a (µg/g)	Predicted Hg Conc in Bluegill Tissue		
		Average (µg/g)	Maximum (µg/g)	Minimum (µg/g)
1988-1989	0.000005	0.037	0.055	0.025
1986-1987	0.000004	0.032	0.047	0.021
1984-1985	0.000005	0.037	0.055	0.025
1982-1983	0.000005	0.039	0.057	0.026
1980-1981	0.000006	0.042	0.062	0.028
1976-1979	0.000006	0.045	0.067	0.030
1973-1975	0.000008	0.060	0.089	0.040
1969-1972	0.000010	0.076	0.112	0.051
1965-1968	0.000013	0.094	0.138	0.063
1962-1964	0.000017	0.127	0.187	0.085
1958-1961	0.000025	0.187	0.275	0.125
1955-1957	0.000043	0.322	0.473	0.215
1952-1954	0.000004	0.027	0.039	0.018
1948-1951	0.000002	0.012	0.017	0.008
1945-1947	0.000001	0.010	0.015	0.007

^a Dissolved Hg, estimated using a particle-to-water distribution ratio (K_d) of 10^5 .

Other mercury data from Watts Bar Reservoir, Clinch River, Poplar Creek, and East Fork Poplar Creek:

Although a large amount of information is available on mercury in fish and sediment from the Clinch and Tennessee River systems, most of the data are of recent vintage; and only one parameter, either water, fish, or sediment, is usually measured in a particular study. However, some of these data can be used to evaluate the predicted mercury concentrations in fish given in Tables 3, 4, 5, and 6. Supplemental data on mercury in fish and sediments from other areas of the Tennessee River system are given in Tables A3 and A4.

The concentrations of mercury in bluegill in East Fork Poplar Creek, Poplar Creek, Clinch River, and Watts Bar Reservoir decrease with distance from Poplar Creek (Elwood 1984, Loar 1981, TVA 1985). This trend is illustrated in Figure 2 which shows data collected for the present study in 1989 and bluegill data collected by Southworth (personal communication) in 1988 from the lower Clinch River, Poplar Creek, and East Fork Poplar Creek. Figure 3 shows the Watts Bar Reservoir data and Southworth's data for the Clinch River, Melton Hill Reservoir, and Hinds Creek, a reference stream that empties into Melton Hill Reservoir. Of interest is the concentration of mercury in bluegill from Hinds Creek. The concentration of mercury in bluegill from this stream, which has no known anthropogenic sources of mercury, is higher than the concentration of mercury in bluegill from Watts Bar Reservoir and Melton Hill Reservoir. Background levels of mercury in the environment will be discussed later.

Elevated levels of mercury in bluegill still exist in EFPC and Poplar Creek below the confluence with EFPC. The average concentration of mercury in bluegill below the Y-12 settling pond (New Hope Pond) had decreased from an average of 2.13 $\mu\text{g/g}$ in 1982 (Van Winkle et al. 1984) to an average of 0.80 $\mu\text{g/g}$ in 1988 (Loar 1989). However, near the mouth of EFPC (mile 1.3) the mean concentration of mercury (37 $\mu\text{g/g}$) in bluegill collected in 1988 (Loar 1988) was slightly greater than the concentration (32.5 $\mu\text{g/g}$) measured in 1982 (Van Winkle et al. 1984). In Poplar Creek below the confluence with

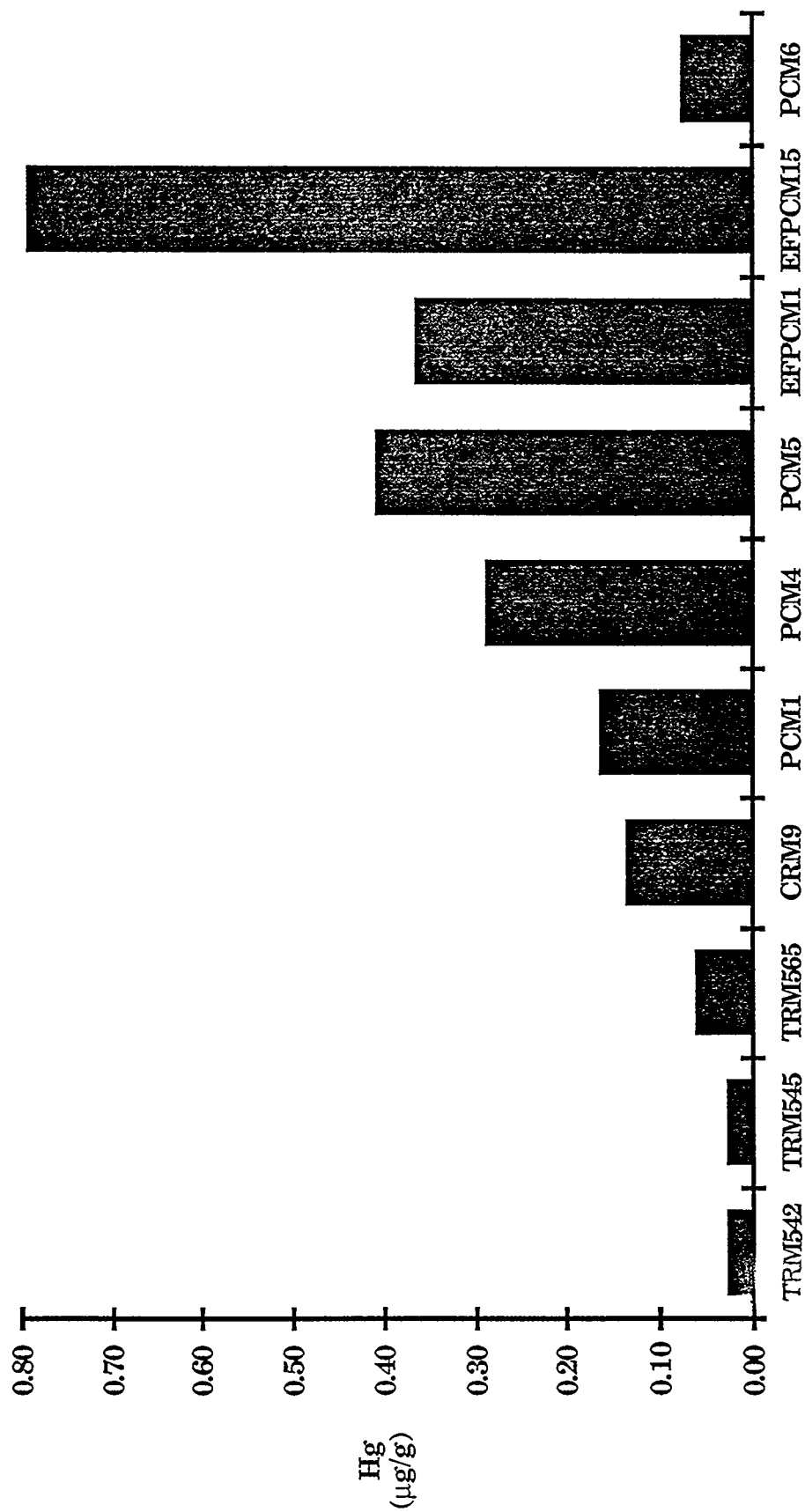


Figure 2. Concentration of mercury in tissue of bluegill (*Lepomis macrochirus*) from Watts Bar Reservoir, Clinch River, Poplar Creek, and East Fork Poplar Creek.

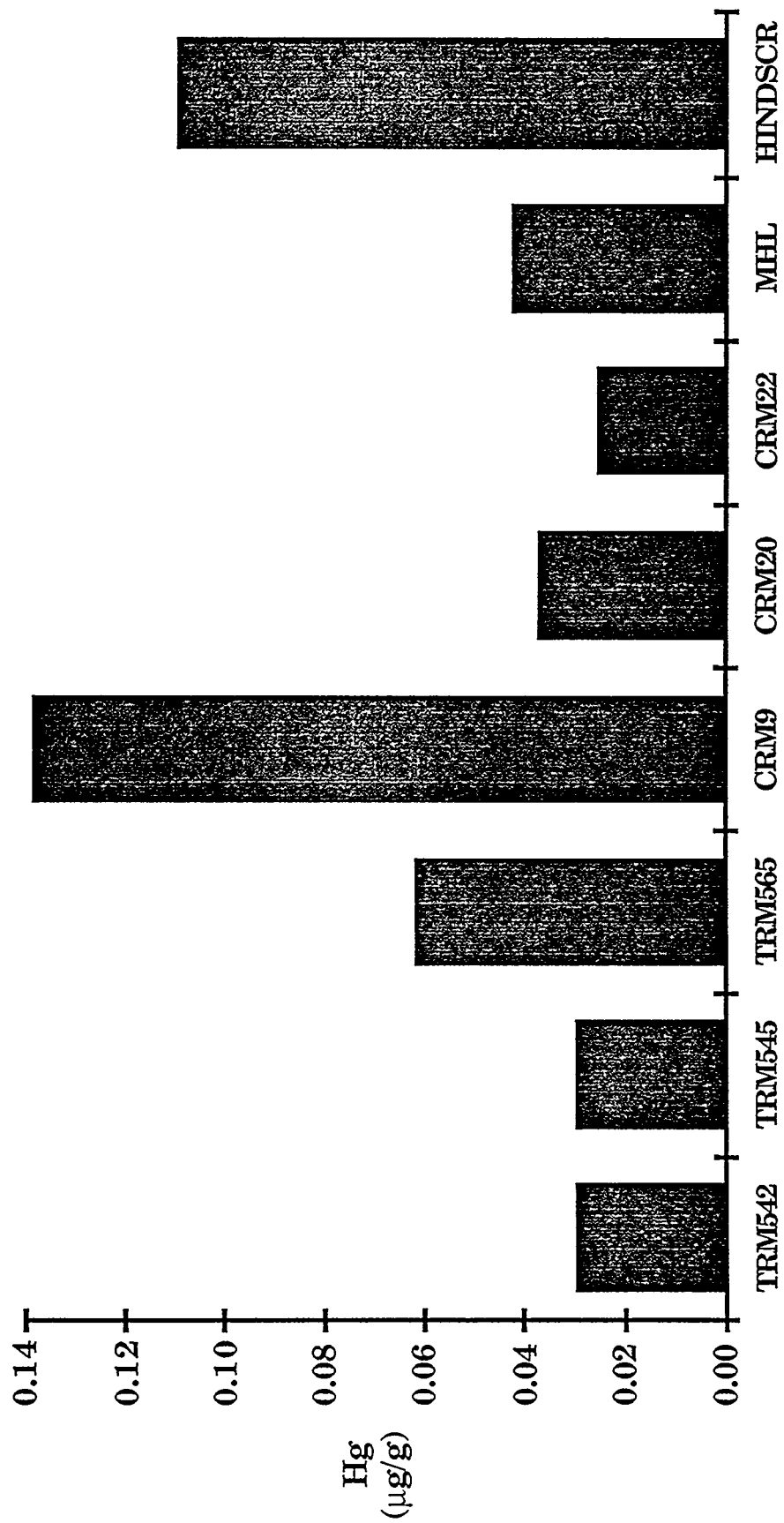


Figure 3. Concentrations of mercury in tissue of bluegill from Watts Bar Reservoir, Clinch River, Melton Hill Reservoir, and Hinds Creek.

EFPC, the mean concentration reported for 1988 by Southworth (Loar 1988) was 0.41 $\mu\text{g/g}$ while in 1976 the average concentration in bluegill for this location was 0.40 $\mu\text{g/g}$ (Elwood 1984). From these selected data and the data shown in Figures 2 and 3, it appears that EFPC is the primary source of mercury to the Clinch River and that mercury concentrations in bluegill in EFPC and the lower end of Poplar Creek have remained fairly constant for many years. The predicted mercury concentrations for bluegill in Watts Bar Reservoir also show little change over the past few years.

Comparison of predicted and measured concentrations of mercury in bluegill in Watts Bar Reservoir

Our predicted concentrations of mercury in bluegill in Watts Bar Reservoir at TRM 536, 545, and White Creek mile 1.3 (~TRM 545) for previous years can be tested by comparing the predicted values with the few measured values that are available for bluegill for previous years. Our average predicted values for these locations ranged from 0.038 to 0.045 $\mu\text{g/g}$ for bluegill in 1984 (Tables 3, 4, and 6) which are in good agreement with TVA's (1985) average measured concentration of 0.04 $\mu\text{g/g}$ of mercury for bluegill collected at TRM 558 during 1984. In 1970, the measured concentration of mercury in bluegill tissue at TRM 555 was <0.050 $\mu\text{g/g}$ (TVA 1972). Average mercury predictions from the current study for 1970 at the two sites near TRM 545 were 0.060 and 0.090 $\mu\text{g/g}$ (Tables 3 and 4). Although the number of comparisons of measured values with predicted values is limited, the agreement between measured and predicted values is very good.

Concentration factors and concentration ratios for mercury from other environments:

Mercury in the North Fork Holston River, Saltville, VA

The North Fork Holston River originates 77 km above the site of a former chloralkali plant and flows 133 km beyond the site where it joins the South Fork Holston River. The two rivers unite to form the Holston River which flows an additional 50 km to Cherokee Reservoir, a man-made reservoir created in 1942. From 1952 until 1972, a chloralkali plant operated near the

bank of the North Fork of the Holston River at Saltville, VA (Hildebrand et al. 1980). During plant operation, salt wastes containing mercury were placed in two disposal ponds adjacent to the river. An estimated 39 kg of mercury leached from the ponds each year. In 1975, concentrations of mercury in sediment downstream from the site were typically 20 times higher than concentrations upstream. Fish species collected progressive distances downstream from the chloralkali plant contained elevated concentrations of mercury which decreased with distance from the plant. Most of the mercury measured in fish tissue was methylmercury (up to 92%).

Data collected in 1974 and 1975 above and below the waste disposal ponds are given in Table 7. Bluegill were not available at the river sites; thus, concentrations of mercury are given for rock bass (*Ambloplites rupestris*). The rock bass is a sunfish that is about the same size as bluegill and belongs to the same family (Centrarchidae). However, rock bass are probably more piscivorous in their feeding habits than bluegill.

Above the disposal ponds the river appears to be uncontaminated with mercury. Concentrations in sediment were less than 1.0 $\mu\text{g/g}$ and the concentration of mercury in water was 0.001 $\mu\text{g/L}$. Concentrations of mercury in fish were relatively high (0.29-0.32 $\mu\text{g/g}$), a situation which results in high CF and CR values. Below the outfall of the disposal ponds, sediments were contaminated with up to 20 $\mu\text{g/g}$ of mercury and concentrations in fish exceeded 1.0 $\mu\text{g/g}$.

The CF for mercury in rock bass collected above the former chloralkali plant is much higher than the CF for bluegill from Watts Bar Reservoir. The relatively high concentrations of mercury in fish above the plant is difficult to explain. Because of the relatively low concentrations of mercury in sediments at this location, the CR values were >1 . Below the former chloralkali plant, the CF values are higher than the ones determined for Watts Bar Reservoir bluegill except for fish collected 3.7 miles below the

Table 7. Concentration of mercury in rock bass tissue, sediment, and water from North Fork Holston River.

Date	Km Above (-) ^a or Below Plant	Rock Bass ^b Hg Conc (µg/g)	Sediment ^c Hg Conc (µg/g)	Water ^d Hg Conc (µg/g)	Fish/ Sediment CR	Fish/ Water CF	Sed/ Water K _d
1975	-43	0.291	0.135	0.000001	2.16	291000	14985
1975	-9	0.322	0.165		1.95		
1975	3.1		18.6	0.00006			30815
1974	3.7	1.623	20.2	0.000186	0.08	8726	108602
1975	9.7	1.814	9.2	0.00004	0.20	45350	230000
1975	21		4.3	0.000027			159259
1975	43	1.292	2.45	0.000027	0.53	47852	10072
1975	133	1.122	0.92	0.000018	1.22	62333	5673

^a (-) Above chloralkali plant

^b Fish values are based on wet wt.

^c Sediment values are based on dry wt.

^d Water values are for dissolved Hg.

Not all samples were taken on same date.

former plant site. At this location the CF and CR values are similar to the site-specific (TRM 545) values determined for Watts Bar Reservoir bluegill.

High CF values for mercury are known to occur in aquatic environments in which the release of mercury has been shut off. Fish previously exposed to high concentrations of mercury will maintain a high body burden because of the long biological half life of mercury in fish. In such environments, after the source of mercury has been eliminated and the water concentration has decreased, the high body burden in fish will result in a high CF value.

Cherokee Reservoir

Cherokee Reservoir and the chloralkali plant at Saltville, VA are somewhat analogous to Watts Bar Reservoir and the Y-12 plant. However, Cherokee Reservoir is farther downstream (more than 100 miles) than Watts Bar Reservoir from the source of mercury contamination. Concentrations of mercury measured in bluegill from Cherokee Reservoir in 1974 and 1975 (Table 8) were about three times higher than the predicted concentration of mercury for bluegill in Watts Bar Reservoir for the same years, except for the site at TRM 567.5 where the concentrations were similar (Tables 3, 4, 5, and 6). Concentration factors for Cherokee Reservoir bluegill were about three times higher than the site-specific CF determined for TRM 545. Concentration ratios were also about a factor of three higher for Cherokee Reservoir than those for bluegill in Watts Bar Reservoir.

Wabigoon-English River System, Ontario, Canada

The Wabigoon River in northwestern Ontario, Canada, leaves Wabigoon Lake at the town of Dryden and flows through Clay Lake before joining the English River approximately 140 km (~87 miles) downstream at Ball Lake. Between 1962 and 1969 approximately ten metric tons of mercury were released into the Wabigoon River from a mercury cell chloralkali operation at Dryden (Parks et al. 1984). Fish sampled from Clay Lake, approximately 80 km (50 miles) below the plant site, were observed to contain mercury concentrations of up to 10 µg/g. Approximately 85% of the mercury in the

Table 8. Concentration of mercury in bluegill, sediment, and water from Cherokee Reservoir.

Date	Km Below Plant	Bluegill ^a Hg Conc ($\mu\text{g/g}$)	Sediment ^b Hg Conc ($\mu\text{g/g}$)	Water ^c Hg Conc ($\mu\text{g/g}$)	Fish/ Sediment CR	Fish/ Water CF	Sed/ Water K_d
1974	196	0.325					
1975	196	0.212	0.81	0.00001	0.26	21200	81000
1974	277	0.155					
1975	277	0.1	0.475	0.000005	0.21	20000	10545

^a Fish values are based on wet wt of tissue.

^b Sediment sample collected 188 Km below plant and values are based on dry wt.

^c Water samples collected 191 Km below plant and values are for dissolved Hg.

edible portion of the fish was methylmercury. In 1970, measures were taken to curtail the release of mercury from the plant; and in 1975, the mercury cells were dismantled. After the mercury cells were dismantled, a two-year study beginning in late 1978 was undertaken to help find methods for alleviating the long-term problem of mercury contamination.

Concentrations of mercury in samples of yellow perch (*Perca flavescens*), sediment, and water from the Wabigoon-English River system collected during 1979-1981 are listed in Table 9. Concentration factors, CRs, and K_d values for Clay Lake are also given; however, the CF and K_d values were calculated using total mercury in the water instead of the dissolved fraction. In many environments such as the Wabigoon-English River system in which the amount of suspended solids is relatively low, the difference between total and dissolved mercury in water is insignificant.

Concentration factors for the Wabigoon-English River system ranged from approximately 2,800 to 25,000. In Clay Lake, the CFs for perch were similar to those for bluegill in Cherokee Reservoir but greater than the CF of 7,500 determined for bluegill in Watts Bar Reservoir. Concentration ratios from noncontaminated areas in the Wabigoon-English River system were >1 which is consistent with the data in Table 7 from the noncontaminated reach of the North Fork Holston River above the former chloralkali plant.

Data from these other environments show that CFs and CRs can vary considerably from environment to environment. Variability in the CF appears to be related to the past history of the mercury release to the environment. In addition, many other factors can influence the CF and CR values. For example, the use of the total mercury in water instead of the dissolved fraction can influence the CF if the amount of suspended solids is high. Sediment type, water quality parameters, and other abiotic as well as biotic factors such as feeding habits, age, etc. can influence the CFs and CRs of fish.

Table 9. Concentration of mercury in perch, sediment, and water collected from the Wabigoon/English River system during 1979-1981.

Location	Sample Type ^a	Hg Conc ^b	Fish/Water CF	Fish/Sed CR	Sed/Water K _d
Wabigoon Lake (upstream)	Perch	0.02	2857	2.0	1429
	Water	7.0			
	Sediment	0.01			
Eagle River (downstream tributary)	Perch	0.06	20000	1.5	13333
	Water	3.0			
	Sediment	0.04			
Clay Lake Inflow ~80 km downstream	Perch	0.42	16154	0.18	92308
	Water	26.0			
	Sediment	2.4			
Clay Lake Outflow ~95 km downstream	Perch	0.4	25000	0.14	175000
	Water	16.0			
	Sediment	2.8			
Wabigoon R inflow to Ball Lake	Perch	0.34	24286	0.37	65714
	Water	14.0			
	Sediment	0.92			
English River inflow to Ball Lake (~140 km)	Perch	0.04	8000	0.571	14000
	Water	5.0			
	Sediment	0.07			
English River outflow of Ball Lake	Perch	0.09	11250	0.375	30000
	Water	8.0			
	Sediment	0.24			

^a All sediment samples are 0-5 cm layer.

^b Units are µg/g for fish and sediment, ng/L for water.

A pattern that appears in these data, but is not always consistent, is that the CRs in uncontaminated streams in which the sediment concentration is very low appear to be much greater than the CRs in contaminated streams (Tables 7 and 9). Another interesting observation is that bluegill from Hinds Creek, an uncontaminated stream, had higher mercury concentrations than bluegill in the downstream lakes (Figure 3).

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Background levels of mercury:

Mean background concentrations reported for mercury in fish range from 0.2 to 0.002 $\mu\text{g/g}$ (Wallace et al. 1971, Harriss 1971). Natural background concentrations of mercury are difficult to determine because few areas are free from all sources of mercury pollution. Even in areas distant from pollution sources, mercury can be introduced into the environment through the impingement of mercury vapor from the burning of mercury-containing coal. In order to obtain data at or near natural background levels of mercury in fish, Huckabee et al. (1974) collected 198 fish consisting of five species from the Great Smoky Mountains National Park in 1972 and analyzed their tissue for mercury content. To confirm the 1972 data, Huckabee et al. (1974) collected fish from these same streams again in 1973. The mean concentrations for all species collected in 1972 and 1973 were 0.035 and 0.036 $\mu\text{g/g}$, respectively. In one of the streams, the concentration of mercury varied from 0.44 $\mu\text{g/g}$ in rainbow trout (*Salmo gairdneri*) to 0.19 $\mu\text{g/g}$ in brook trout (*Salvelinus fontinalis*). These data probably provide the best background level for mercury that is available for fish in this area.

A comparison of the predicted values for bluegill in Watts Bar Reservoir (Tables 3, 4, 5, and 6) prior to 1950 gave values for mercury that ranged from 0.075 to 0.015 $\mu\text{g/g}$. This value is in relatively good agreement with the value of 0.035 $\mu\text{g/g}$ reported by Huckabee et al. (1974) for background levels in fish. This agreement of predicted and background values is supportive of the method used in the current study to predict the concentration of mercury in fish for previous years. The higher predicted concentration of mercury in fish at TRM 567.5 from 1946 to 1951 than at the other locations in Watts Bar Reservoir is related to the higher concentrations of mercury in

sediment at this site and, as discussed previously, can be attributed to the influence of the Clinch River. Although large quantities of mercury were not released from the Y-12 plant until 1955, the Oak Ridge facilities began operating in 1944; and mercury is a common pollutant from many industrial facilities.

Discussion:

It is evident from the data presented from environments other than Watts Bar Reservoir that CFs can fluctuate greatly. Often these differences are due to the method used to calculate the CF (e.g., whole fish vs muscle, wet wt vs dry wt, etc.). Probably the most important factor is whether mercury in the fish is in equilibrium with mercury in the environment. In aquatic environments in which the source of mercury has been eliminated, fish that retain their body burden of mercury can have very high CF values because of the rapid decrease of mercury in water and the long biological half-life of mercury in fish. This appears to be the situation for the Wabigoon-English River system and the North Fork Holston River.

A major shortcoming of the present study is that in Watts Bar Reservoir, measurements of all three parameters: water, sediment, and fish, were available for only the TRM 545 site (Table 2). Additional spatial and temporal measurements of these three parameters and more past fish data for testing our predictions would increase the confidence in our predicted values. However, for the few predictions that could be tested against measured data for previous years, the predictions were extremely good.

Estimated health risk to humans as a result of consuming bluegill from Watts Bar Reservoir:

To estimate the health risk associated with consuming bluegill from the lower end of Watts Bar Reservoir, it is necessary to know the level that mercury would reach in the tissues of an individual who regularly consumed these fish. Mercury is ubiquitous in the environment and everyone's tissues contain mercury. We are constantly taking mercury into our bodies through the daily consumption of food and liquids. In 1973, the

Food and Drug Administration (FDA) indicated an average dietary intake of 2.89 $\mu\text{g/day}$ of mercury. Most of the intake was from fish (Food Chemical News: August 4, 1975). Although data are scarce, the "normal" intake of mercury by humans has been estimated to be about 3 to 4 $\mu\text{g/day}$ (Gesterner and Huff 1977).

The level of mercury that accumulates in human tissues depends on many factors such as the amount and form taken in and the biological half-life in man. The biological half-life is the time required for the mercury concentration in a person to decrease by one-half, providing no additional mercury is taken into the body. The biological half-life reported for methylmercury in humans is 76 ± 3 days (World Health Organization 1976). The biological half-life varies from person to person and other data have suggested a biological half-life of about 70 days. Following a single intake, methylmercury would be absorbed from the gut and distributed to various body tissues. Half of the mercury would be lost in about 70 days and only about 1.5% would remain after 420 days.

For repeated intakes, an equilibrium level is reached between intake into and loss from the body. The equilibrium body level for mercury is about 100 times intake. For small daily intakes the equilibrium level will be small; accordingly, for large daily intakes the equilibrium level will be large. In either case, the equilibrium level in the body, sometimes called body burden, will be about 100 times the daily intake.

Methylmercury is the chemical form of mercury that is the most toxic to humans. Usually about 80 to 95% of the total mercury in fish is reported to be methylmercury, and the biological half-life is usually longer in fish than in man. In order to estimate the health risk to humans from consuming bluegill from the lower end of Watts Bar Reservoir, we will assume that all mercury in the fish is methylmercury and that when bluegill are eaten by humans, all of the methylmercury in the fish tissue is absorbed through the human gastrointestinal tract.

It is necessary to know the amount of bluegill tissue consumed and the frequency at which the fish are consumed to estimate the concentration of mercury that will be reached in the tissue of humans. Rupp (1980) reviewed surveys of fish consumption in the U.S. and reported that among adults who eat fish, the average consumption of freshwater fish for U.S. residents is 3.41 kg/yr (9.3 g/day).

From studies conducted in Sweden, Finland, and Japan (Federal Register 1979), it was concluded that the lowest blood level of mercury that would bring about signs and symptoms of methylmercury poisoning was 200 parts per billion (ppb) (0.200 $\mu\text{g/g}$). The body burden for an average adult man weighing 70 kg would be approximately 30,000 μg (30 mg) of mercury. To obtain this equilibrium level requires a daily intake of 300 μg of methylmercury. In setting standards, a safety factor of 10 is usually applied. Thus a maximum tolerable level would be 20 ppb (0.020 $\mu\text{g/g}$) of methylmercury in blood or 30 μg of methylmercury daily in the diet. After the promulgation by the FDA of an action level of 0.5 $\mu\text{g/g}$ for unavoidable residues of mercury in the edible portion of fish and shellfish, additional data on mercury in humans resulted in the action level being increased to 1.0 $\mu\text{g/g}$ (Federal Register 1979). Using the average adult consumption rate of 9.3 g/day for freshwater fish reported by Rupp (1980), an action level of 1.0 μg of mercury per gram of fish would provide a daily intake of 9.3 μg of mercury.

In a screening analysis conducted on identified contaminants in EFPC, Hoffman et al. (1984) used an acceptable daily intake (ADI) of 20 μg for methylmercury. Lappenbusch (1988) reviewed the EPA's recommendations and found a reference dose (RfD) of 3×10^{-4} mg/kg/day for the oral intake of methylmercury from which an oral ADI of 21 μg for a 70-kg adult can be calculated. Thus, the ADIs are much less than the intake of 300 $\mu\text{g/day}$ required to produce signs and symptoms of methylmercury poisoning (Federal Register 1979).

To determine whether the routine consumption of bluegill from the lower end of Watts Bar Reservoir at any time during the past four decades posed a human health risk, we need to know the average daily intake of mercury. We will assume that all mercury in bluegill tissue is methylmercury and that it is totally absorbed through the human gastrointestinal tract. In addition, we will use data from the time period and location when mercury concentrations were highest.

The highest predicted concentrations of mercury in bluegill occurred during the period between 1956 and 1960. If the estimated concentration of mercury in bluegill at each site is averaged for this five-year period, the highest average concentration would be 1.5 $\mu\text{g/g}$ at TRM 567.5. A five-year average is used because, as the data in Table A1 show, most bluegill would take about 5 years to reach an acceptable size for eating. An average consumption rate of 9.3 g/day of fish (Rupp 1980) and a mercury concentration of 1.5 $\mu\text{g/g}$ provide an average daily intake of 14.0 μg of mercury. Although the 1.5 $\mu\text{g/g}$ estimated concentration of mercury in bluegill from TRM 567.5 exceeds the FDA action level of 1.0 $\mu\text{g/g}$ for fish, the daily intake for an average adult is less than the ADI of 20 μg used by Hoffman et al. (1984) in his screening analysis and less than 5% of the 300 $\mu\text{g/day}$ needed to produce signs and symptoms of mercury poisoning (Federal Register 1979).

Fish-eating habits, especially freshwater fish, vary from one region of the U.S. to another. Rupp (1980) reported regional consumption rates and a maximum for each region. Tennessee is included in the East South Central region and the maximum consumption rate for adults surveyed in this region during 1973-1974 was 24.64 kg/yr (67.5 g/day). The maximum consumer would be considered the "critical" individual, i.e., the person with the greatest potential for being affected by contaminants contained in fish tissue. If the maximum consumption rate is applied to bluegill from Watts Bar Reservoir containing 1.5 μg of mercury per gram of tissue, then the intake of mercury would be 101.25 $\mu\text{g/day}$. This quantity is approximately five times the recommended maximum daily intake but is

only one-third the level at which signs and symptoms of mercury poisoning are evident (Federal Register 1979). The conclusion from the present analysis is that during 1956-1960 when the predicted concentrations of mercury in bluegill were the highest, the probability of mercury poisoning occurring as a result of eating bluegill from the lower end of Watts Bar Reservoir was very low. However, because mercury can be taken into the body by other dietary pathways as well as by inhalation, high concentrations in fish must be considered a potential health risk.

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APPENDIX

Table A1. Length, weight, and mercury concentration in bluegill collected March 9, 1989 from Watts Bar Reservoir.

DATE	SAMPLING LOCATION	SPECIES	AGE (years)	ID	FISH LENGTH (cm)	FISH WEIGHT (grams)	TISSUE HG (a) (ng/g)	TISSUE HG (b) (ng/g)	TISSUE Ave Hg (ng/g=ppb)	Stdev	Ave Length
9-Mar-89	TRM 545	Bluegill	5	385	16.0	60.9	20	21	20.5		
9-Mar-89	TRM 545	Bluegill		386	15.7	64.2	25	26	25.5		
9-Mar-89	TRM 545	Bluegill		387	14.2	44	NS				
9-Mar-89	TRM 545	Bluegill	6	388	18.4	96.2	44	40	42.0		
9-Mar-89	TRM 545	Bluegill	4	389	14.5	44.1	31	30	30.5		
9-Mar-89	TRM 545	Bluegill		370	16.0	66.2	39	42	40.5		
9-Mar-89	TRM 545	Bluegill		371	15.4	55.2	34	30	32.0	8.03	15.61
9-Mar-89	TRM 545	Bluegill		372	17.0	83.6	28	30	29.0		
9-Mar-89	TRM 545	Bluegill		373	15.5	61.8	NS			Ave Hg	Ave Wt
9-Mar-89	TRM 545	Bluegill		374	13.4	33.6	22	20	21.0	30.13	60.98
9-Mar-89	TRM 542	Bluegill		375	14.9	52.7	54	42	48.0		
9-Mar-89	TRM 542	Bluegill	4	376	15.0	55.8	17		17.0	Stdev	Ave Length
9-Mar-89	TRM 542	Bluegill	5	377	15.0	56.7	36		36.0	8.97	14.925
9-Mar-89	TRM 542	Bluegill		378	15.0	54.9	29		29.0	Ave Hg	Ave Wt
9-Mar-89	TRM 542	Bluegill		379	14.5	49.9	31		31.0	30.25	53.075
9-Mar-89	TRM 542	Bluegill		380	15.0	54	28		28.0		
9-Mar-89	TRM 542	Bluegill		360	15.0	53.4	25		25.0		
9-Mar-89	TRM 542	Bluegill		361	15.0	47.2	28		28.0		
9-Mar-89	TRM 542	Bluegill		364	13.7	42	NS				
9-Mar-89	TRM 542	Bluegill		365	13.0	35	NS				
9-Mar-89	TRM 565	Bluegill		399	16.0	66.8	89	83	86.0		
9-Mar-89	TRM 565	Bluegill		398	15.5	58.4	69		69.0	Stdev	Ave Length
9-Mar-89	TRM 565	Bluegill		397	15.5	61.1	125		125.0	36.07	15.525
9-Mar-89	TRM 565	Bluegill		396	15.5	62.6	74		74.0	Ave Hg	Ave Wt
9-Mar-89	TRM 565	Bluegill		395	15.7	66.4	30		30.0	62.13	60.225
9-Mar-89	TRM 565	Bluegill		394	15.5	55.6	22		22.0		
9-Mar-89	TRM 565	Bluegill		393	15.5	52.7	70		70.0		
9-Mar-89	TRM 565	Bluegill		392	15.0	58.2	21		21.0		
9-Mar-89	TRM 565	Bluegill		391	16.5	67.1	NS				
9-Mar-89	TRM 565	Bluegill		390	15.5	55.9	NS				

NS = Not submitted

a & b = replicate samples

Table A2. Concentration of mercury in Watts Bar Reservoir sediment cores and predicted concentrations for water and fish.

WATTS BAR SEDIMENT CORE 8-2-3 (Oct 17, 1986) APPROX TRM 545													
CORE DEPTH (cm)	SAMPLE DRY WT. (gm)	CS-137 IN SEDIMENT (pCi/g)	2 sigma Counting Error	HG IN SEDIMENT (ppm)	DISSOLVED HG IN H ₂ O* (ppb)	HISTORICAL** TIME-PERIOD (Years)	PRED BG (ppb) USING GAVE WATERCF	PRED BG (ppb) USING MAX WATERCF	PRED BG (ppb) WATERCF	PRED BG (ppb) USING GAVE SEDCR	PRED BG (ppb) USING MAX SEDCR	PRED BG (ppb) USING MIN SEDCR	PRED BG (ppb) USING MAX SEDCR
0-4	13.81	3.74	0.10	0.52	0.005	1984-1986	38	55	25	30	44	20	20
4-8	18.10	3.80	0.10	0.53	0.005	1981-1983	38	55	25	31	45	20	20
8-12	26.93	3.71	0.33	0.52	0.005	1978-1980	38	55	25	30	44	20	20
12-16	31.17	4.43	0.24	0.64	0.006	1975-1977	45	66	30	37	54	24	24
16-20	31.26	4.22	0.30	0.72	0.007	1972-1974	53	77	35	42	61	27	27
20-24	32.77	5.19	0.25	0.78	0.008	1969-1971	60	88	40	45	66	30	30
24-28	37.01	5.84	0.35	1.07	0.011	1966-1968	83	121	55	62	91	41	41
28-32	39.95	11.76	0.40	1.75	0.018	1964-1965	135	198	90	102	149	67	67
32-36	43.53	15.87	0.53	2.30	0.023	1961-1963	173	253	115	133	196	87	87
36-40	44.33	24.34	0.47	4.69	0.047	1958-1960	353	517	235	272	399	178	178
40-44	42.08	27.57	0.50	5.46	0.055	1956-1958	413	605	275	317	464	207	207
44-48	42.64	9.45	0.42	1.08	0.011	1953-1955	83	121	55	63	92	41	41
48-52	46.06	10.53	0.30	0.47	0.005	1950-1952	38	55	25	27	40	18	18
52-56	48.91	11.67	0.41	0.29	0.003	1947-1949	23	33	15	17	25	11	11
56-60	63.41	3.33	0.19	0.14	0.001	1944-1946	8	11	5	8	12	5	5
60-62	53.50	0.29	0.07	0.06	0.001		8	11	5	3	5	2	2

* Estimated using a particle-to-water distribution ratio of 10E5

** Estimated using a sediment accumulation rate of 1.4 cm/yr

Table A2 (cont.)

WATTS BAR SEDIMENT CORE TRM 567.5 (AUG 22, 1986)													
CORE DEPTH (cm)	SAMPLE DRY WT. (gm)	SEDIMENT (pCi/g)	CS-137 IN 2 sigma Counting Error	HG IN SEDIMENT (ppm)	DISSOLVED HG IN H ₂ O* (ppb)	HISTORICAL** TIME PERIOD (Years)	PRED BG (ppb) USING AVE WATERCF	PRED BG (ppb) USING MAX WATERCF	PRED BG (ppb) WATERCF	PRED BG (ppb) USING AVE SEDCR	PRED BG (ppb) USING MAX SEDCR	PRED BG (ppb) USING AVE SEDCR	PRED BG (ppb) USING MAX SEDCR
0-4	72.58	5.26	0.06	1.52	0.015	1986	113	165	75	62	125	62	21
4-8	119.62	5.68	0.07	1.36	0.014	1984-1985	105	154	70	56	112	56	19
8-12	127.51	5.06	0.08	1.77	0.018	1983	135	198	90	73	145	73	25
12-16	140.85	6.05	0.19	2.42	0.024	1981-1982	180	264	120	99	198	99	34
16-20	147.80	7.39	0.07	3.16	0.032	1980	240	352	160	130	259	130	44
20-24	144.13	6.51	0.06	2.32	0.023	1978-1979	173	253	115	95	190	95	32
24-28	138.34	6.61	0.06	2.32	0.023	1977	173	253	115	95	190	95	32
28-32	109.97	7.85	0.06	3.33	0.033	1975-1976	248	363	165	137	273	137	47
32-36	101.34	11.01	0.09	3.44	0.034	1974	255	374	170	141	282	141	48
36-40	133.20	9.81	0.06	2.98	0.030	1972-1973	225	330	150	122	244	122	42
40-48	144.17	12.03	0.24	2.91	0.029	1969-1971	218	319	145	119	239	119	41
48-56	149.98	13.56	0.11	4.12	0.041	1966-1968	308	451	205	169	338	169	58
56-64	161.15	19.02	0.15	5.10	0.051	1963-1965	383	561	255	209	418	209	71
64-72	156.36	22.24	0.11	6.80	0.068	1960-1962	510	748	340	279	558	279	95
72-80	145.92	49.08	0.21	24.40	0.244	1957-1959	1830	2684	1220	1000	2001	1000	342
80-84	70.62	58.36	0.17	19.00	0.190	1955-1956	1425	2090	950	779	1558	779	266
84-88	78.68	28.66	0.11	6.31	0.063	1952-1954	473	693	315	259	517	259	88
88-92	81.22	13.01	0.07	1.24	0.012	1950-1951	90	132	60	51	102	51	17
92-96	79.15	18.59	0.08	0.36	0.004	1948-1949	30	44	20	15	30	15	5
96-100	77.05	10.64	0.07	0.35	0.004	1946-1947	30	44	20	14	29	14	5
100-104	76.68	13.11	0.07	0.31	0.003								
104-108	75.71	5.84	0.05	0.34	0.003								
108-112	81.48	0.87	0.02	0.12	0.001								
112-116	118.11	0.25	0.03	0.09	0.001								
116-120	119.10	0.27	0.05	0.05	0.001								
120-122	125.07	0.00	0.00										

* Estimated using a particle-to-water distribution ratio of 10E5

** Estimated using a sediment accumulation rate of 2.8 cm/yr

Table A2 (cont.)

WHITE CREEK-WATTS BAR SEDIMENT CORE WCM 1.3 RCB (MAR 22, 1989)
APPROX. TRM 545

CORE DEPTH (cm)	SAMPLE DRY WT. (gm)	CS-137 IN SEDIMENT (pCi/g)	2 sigma Counting Error	HG IN SEDIMENT (ppm)	DISSOLVED HG IN H ₂ O* (ppb)	HISTORICAL** TIME PERIOD (Years)	PRED BG (ppb) USING AVE WATERCF	PRED BG (ppb) USING MAX WATERCF	PRED BG (ppb) USING AVE SEDCR	PRED BG (ppb) USING MAX SEDCR	PRED BG (ppb) USING MIN SEDCR
0-3	19.58	1.89	0.05	0.52	0.005	1988-1989	38	55	30	44	20
3-6	24.79	2.29	0.04	0.55	0.006	1987	45	66	32	47	21
6-9	25.88	2.40	0.04	0.54	0.005	1986	38	55	31	46	21
9-12	31.02	2.71	0.17	0.52	0.005	1985	38	55	30	44	20
12-15	36.27	2.36	0.14	0.55	0.006	1984	45	66	32	47	21
15-20	51.32	3.23	0.15	0.61	0.006	1981-1983	45	66	35	52	23
20-25	56.10	3.17	0.13	0.68	0.007	1979-1980	53	77	39	58	26
25-30	55.86	3.49	0.16	0.72	0.007	1977-1978	53	77	42	61	27
30-35	55.57	4.19	0.16	0.89	0.009	1975-1976	68	99	52	76	34
35-40	57.83	4.63	0.17	0.97	0.010	1973-1974	75	110	56	82	37
40-45	61.85	5.59	0.17	1.14	0.011	1971-1972	83	121	66	97	43
45-50	59.13	7.24	0.20	1.22	0.012	1969-1970	90	132	71	104	46
50-55	60.10	8.40	0.20	1.52	0.015	1967-1968	113	165	88	129	58
55-60	60.59	9.44	0.21	1.56	0.016	1965-1966	120	176	90	133	59
60-65	62.40	12.18	0.23	2.37	0.024	1963-1964	180	264	137	201	90
65-70	63.11	14.05	0.24	2.74	0.027	1961-1962	203	297	159	233	104
70-75	59.87	20.53	0.31	5.00	0.050	1959-1960	375	550	290	425	190
75-80	61.37	25.60	0.33	8.03	0.080	1957-1958	600	880	466	683	305
80-85	64.84	17.41	0.27	6.54	0.065	1955-1956	488	715	379	556	249
85-90	65.09	6.96	0.18	0.40	0.004	1953-1954	30	44	23	34	15
90-95	64.31	8.66	0.20	0.21	0.002	1951-1952	15	22	12	18	8
95-100	57.96	9.83	0.22	0.16	0.002	1949-1950	15	22	9	14	6

* Estimated using a particle-to-water distribution ratio of 10E5

** Estimated using a sediment accumulation rate of 2.4 cm/yr

Table A2 (cont.)

WATTS BAR SEDIMENT CORE TRM 536.5: (MAR 22, 1989)													
CORE DEPTH (cm)	SAMPLE DRY WT. (gm)	CS-137 IN SEDIMENT (pCi/g)	2 sigma Counting Error	HG IN SEDIMENT (ppm)	DISSOLVED HG IN H ₂ O* (ppb)	HISTORICAL** TIME-PERIOD (Years)	PRED BG (ppb) USING AVE WATERCF	PRED BG (ppb) USING MAX WATERCF	PRED BG (ppb) USING MIN WATERCF	PRED BG (ppb) USING AVE SEDCR	PRED BG (ppb) USING MAX SEDCR	PRED BG (ppb) USING MIN SEDCR	
0-3	20.08	1.69	0.06	0.50	0.005	1988-1989	38	55	25	29	43	19	
3-6	21.51	1.74	0.06	0.43	0.004	1986-1987	32	47	22	25	37	16	
6-9	24.86	1.78	0.04	0.50	0.005	1984-1985	38	55	25	29	43	19	
9-12	31.23	1.89	0.20	0.52	0.005	1982-1983	39	57	26	30	44	20	
12-15	34.12	2.13	0.16	0.57	0.006	1980-1981	43	63	29	33	48	22	
15-20	52.14	2.72	0.18	0.61	0.006	1976-1979	46	67	31	35	52	23	
20-25	60.11	4.08	0.20	0.81	0.008	1973-1975	61	89	41	47	69	31	
25-30	58.34	5.28	0.21	1.02	0.010	1969-1972	77	112	51	59	87	39	
30-35	60.21	6.84	0.23	1.26	0.013	1965-1968	95	139	63	73	107	48	
35-40	62.38	9.47	0.26	1.70	0.017	1962-1964	128	187	85	99	145	65	
40-45	66.57	11.99	0.29	2.50	0.025	1958-1961	188	275	125	145	213	95	
45-50	69.08	13.87	0.30	4.30	0.043	1955-1957	323	473	215	249	366	163	
50-55	65.83	6.62	0.21	0.36	0.004	1952-1954	27	40	18	21	31	14	
55-60	76.12	7.88	0.22	0.16	0.002	1948-1951	12	18	8	9	14	6	
60-65	78.23	6.54	0.19	0.14	0.001	1945-1947	11	15	7	8	12	5	
65-70	75.67	1.15	0.10	0.13	0.001								
70-75	96.29	0.17	0.04	0.08	0.001								
75-78	52.23	0.00	0.00	0.06	0.001								

* Estimated using a particle-to-water distribution ratio of 10E5.

** Estimated using a sediment accumulation rate of 1.5 cm/yr